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Publication date:
2015

Document Version
Peer reviewed version

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Citation (APA):

A Razak, A. H., Szabo, P., & Skov, A. L. (2015). *Enhancing relative permittivity by incorporating PDMS-PEG multiblock copolymers in binary polymer blends*. Poster session presented at 5th International Conference on Electromechanically Active Polymer (EAP) Transducers & Artificial Muscles, Tallinn, Estonia.

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Poster ID:

1.2.11

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Enhancing relative permittivity by incorporating PDMS-PEG multi block copolymers in binary polymer blends

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Abstract

Polydimethylsiloxane (PDMS) elastomers are well-known to be soft and highly stretchable, yet they never achieve maximum elongation when utilised as dielectric elastomers, simply because their dielectric permittivity remains rather low. Conversely, polyethyleneglycols (PEG) are not stretchable, but they do possess high permittivity. Combining two such polymers in a block copolymer allows for further cross-linking and presents the possibility of substantial improvements in the actuation response of the resulting dielectric elastomer – if carefully designed. The objective is to synthesise a PDMS-PEG multiblock copolymer, assembling it into discontinuous morphologies in PEG in a continuous PDMS network. PDMS-PEG multiblock copolymers, however, have been shown to be conductive and thus not capable of actuating. By incorporating conductive PDMS-PEG multiblock copolymers into a commercial PDMS elastomer, the discontinuity in PEG can be acquired and the relative permittivity (ϵ_r) is significantly enhanced (60%) with 5wt% of PDMS-PEG block copolymer incorporated into the silicone elastomer.

1. Introduction

The astonishing feature of block copolymers is the variety of morphologies due to self-assembly in bulk or in solution.^{1,2} In principle, a diblock copolymer which is the simplest block copolymer assembles into different morphologies such as sphere (S), cylinder (C), gyroid (G) and lamellar (L).^{1,3} These morphologies can be achieved when two immiscible, covalently bonded polymers microphase separately.⁴ These morphologies can be changed by varying the volume fraction of one constituent in the diblock copolymer. For triblock copolymers, the morphologies are more complex mainly due to the sequence order of three distinct polymers e.g. ABC, ACB, BAC and BCA, which introduces further degrees of freedom and thus allows for the assembly of nearly 30 different morphologies.¹ The similarity shared by the block copolymers is that they have four common equilibrium morphologies (S, C, G and L) (**figure 1**).³

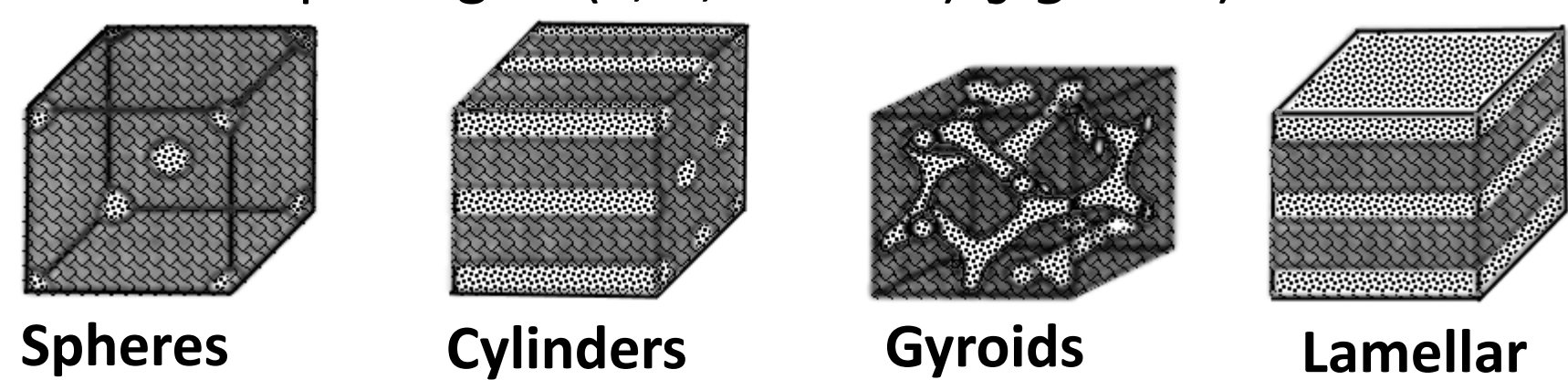
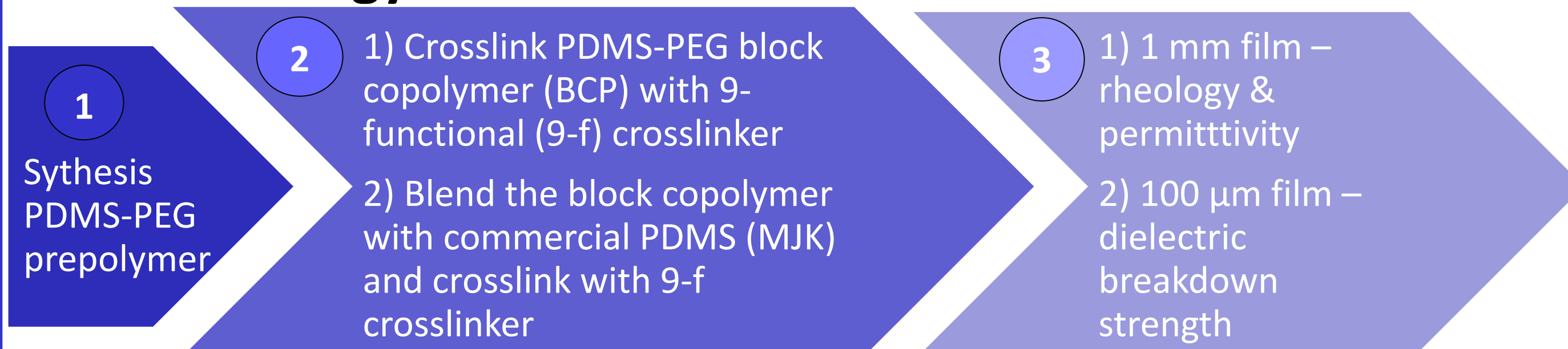


Figure 1: Common morphologies obtained for diblock copolymer when varying volume fraction of one constituent polymer in diblock copolymer ranging from the lowest volume until the highest volume fraction.⁵

2. Methodology



Here, elastomers are prepared by means of phase separating PDMS-PEG multiblock copolymers, whereby the copolymers' blocks are expected to segregate to form very well-defined structures, depending on the chain-lengths of the two constituents (**table 1**). The utilized synthesis of PDMS-PEG multiblock copolymer is based on hydrosilylation reaction with presence of platinum II complex catalyst (**figure 2**).

Table 1: Sample details for PDMS-PEG multiblock copolymer.

PDMS-PEG block copolymer	Number average molecular weight of PDMS (M_n , PDMS) [g/mol]	Number of repeating units in PDMS (m)	Stoichiometric ratio (r)	Volume fraction of PDMS (f_{PDMS})
PDMS81-PEG	6000,00	81	1.21	0.94
PDMS14-PEG	1050,00	14	1.04	0.75
PDMS7-PEG	550,00	7	1.03	0.62
PDMS3-PEG	208,00	3	1.02	0.45

Note: M_n of PEG in PDMS-PEG block copolymer is 250 g/mol

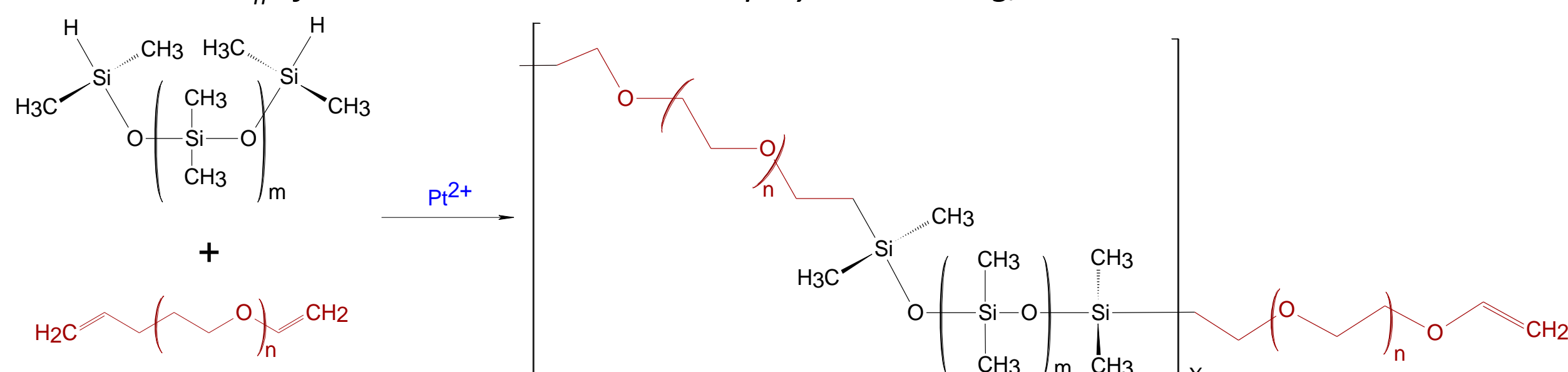
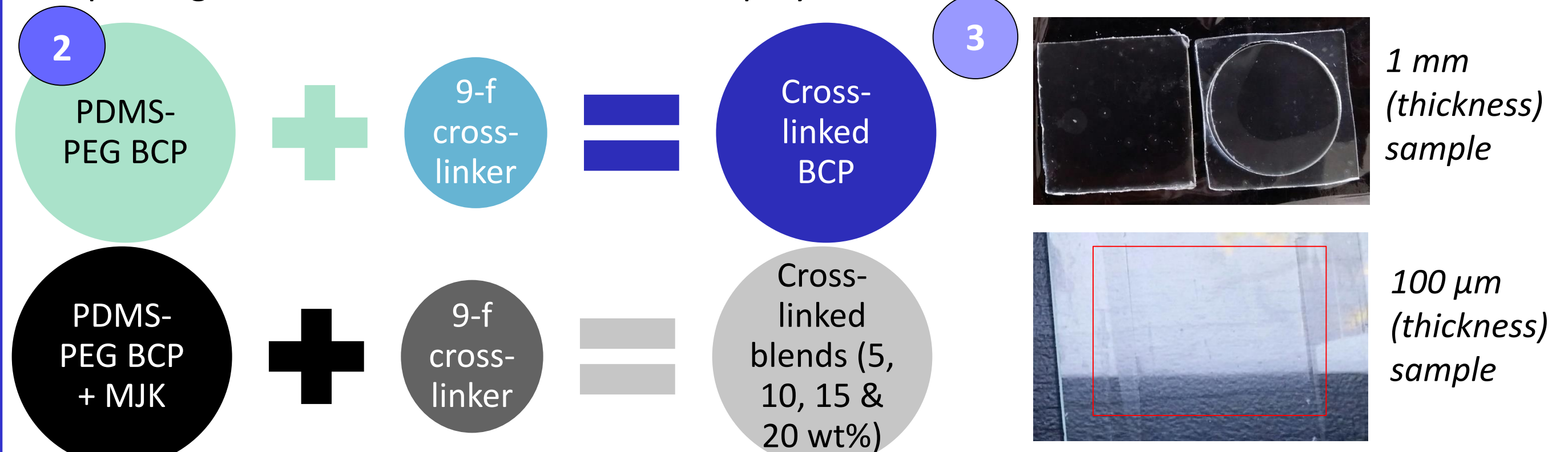


Figure 2: A hydrosilylation reaction when preparing a PDMS-PEG multiblock copolymer with presence of platinum catalyst, where m is the number of repeating dimethylsiloxane units in PDMS, and $n=4$ is the constant number of repeating ethyleneglycol units in PEG. X is the number of repeating PDMS-PEG units in multiblock copolymers.

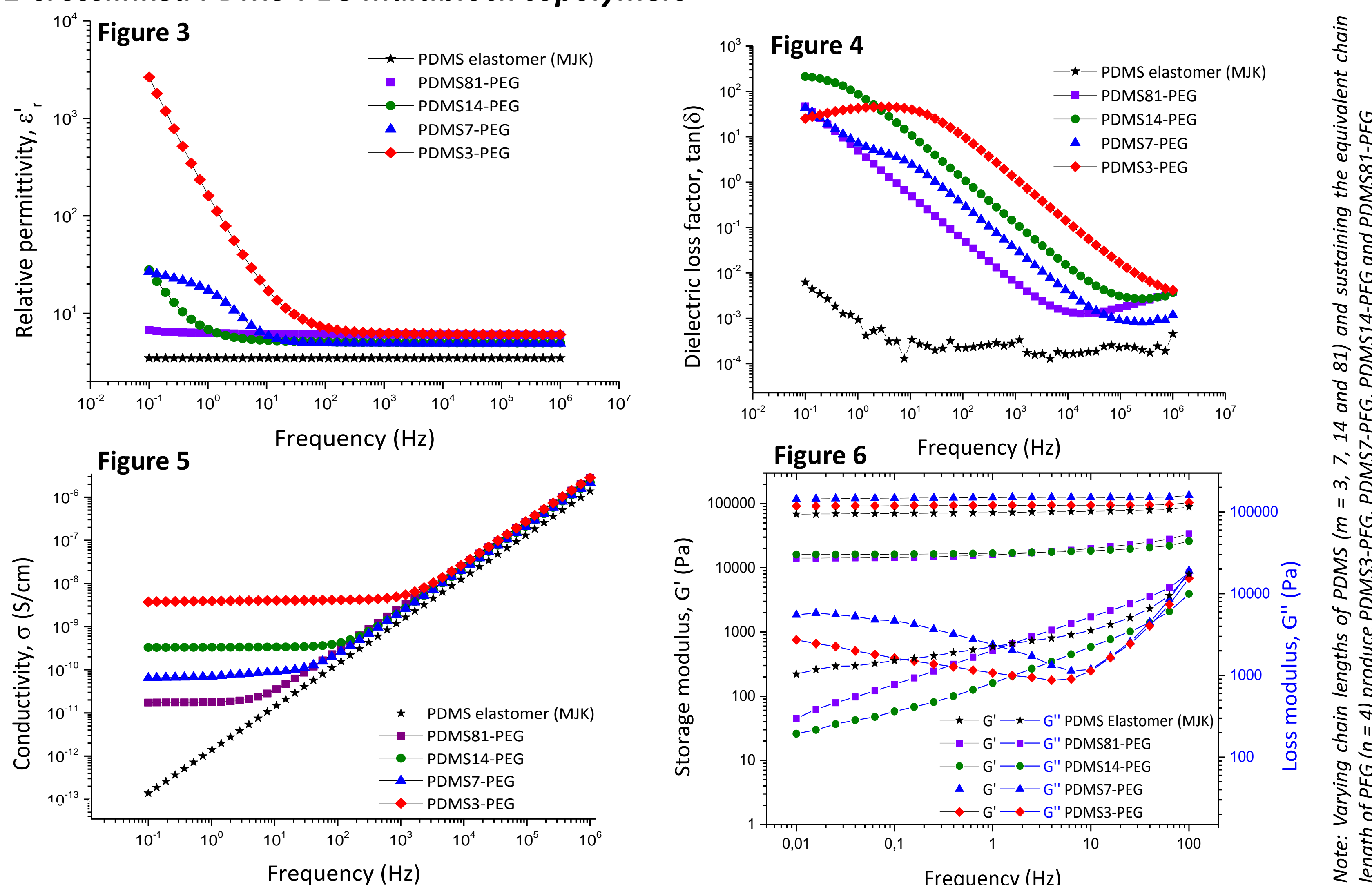


4. Conclusion

A new composite elastomer, which has high relative and low permittivity was successfully created from a binary system of polymer blends consisting of conducting PDMS7-PEG multiblock copolymer and non-conducting PDMS elastomer (MJK). Incorporating 5 wt % of PDMS7-PEG block copolymer into silicone network increases the relative permittivity up to 60% compared to MJK and gives low storage modulus (G') resembling to G' of MJK.

3. Data and Results

3.1 Crosslinked PDMS-PEG multiblock copolymers



Results for PDMS-PEG multiblock copolymers at 23 °C: **Figure 3** Relative permittivity. **Figure 4** Dielectric loss factor **Figure 5** Conductivity. **Figure 6** Storage and loss modulus.

3.2 Crosslinked polymer blends (MJK/PDMS7)

Due to the conductivity of PDMS-PEG multiblock copolymers (**figure 5**), they were blended and cross-linked with commercial PDMS elastomer (MJK) – MJK/PDMS7. Incorporating the block copolymers into silicone network as binary polymer blend (BPB) creates promising morphologies which gives high relative (**figure 8**) and low loss permittivity (**figure 9**) compared to the reference silicone elastomer (MJK).

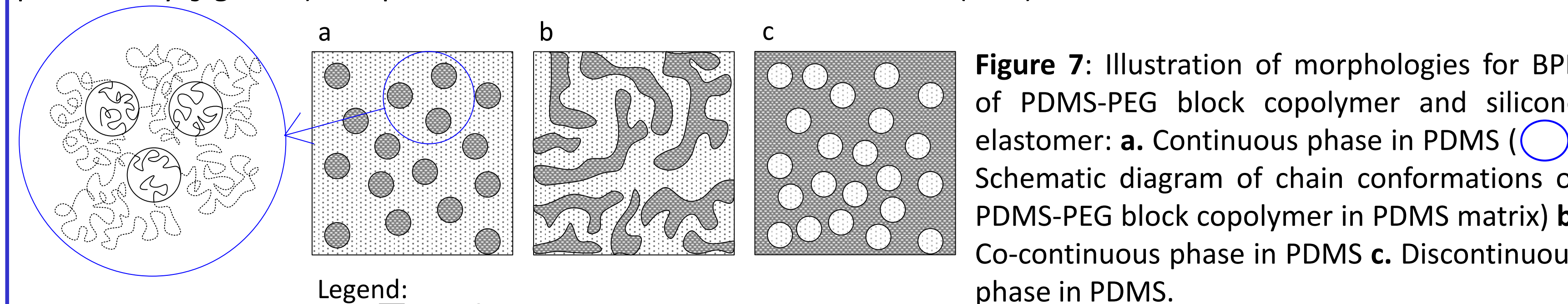
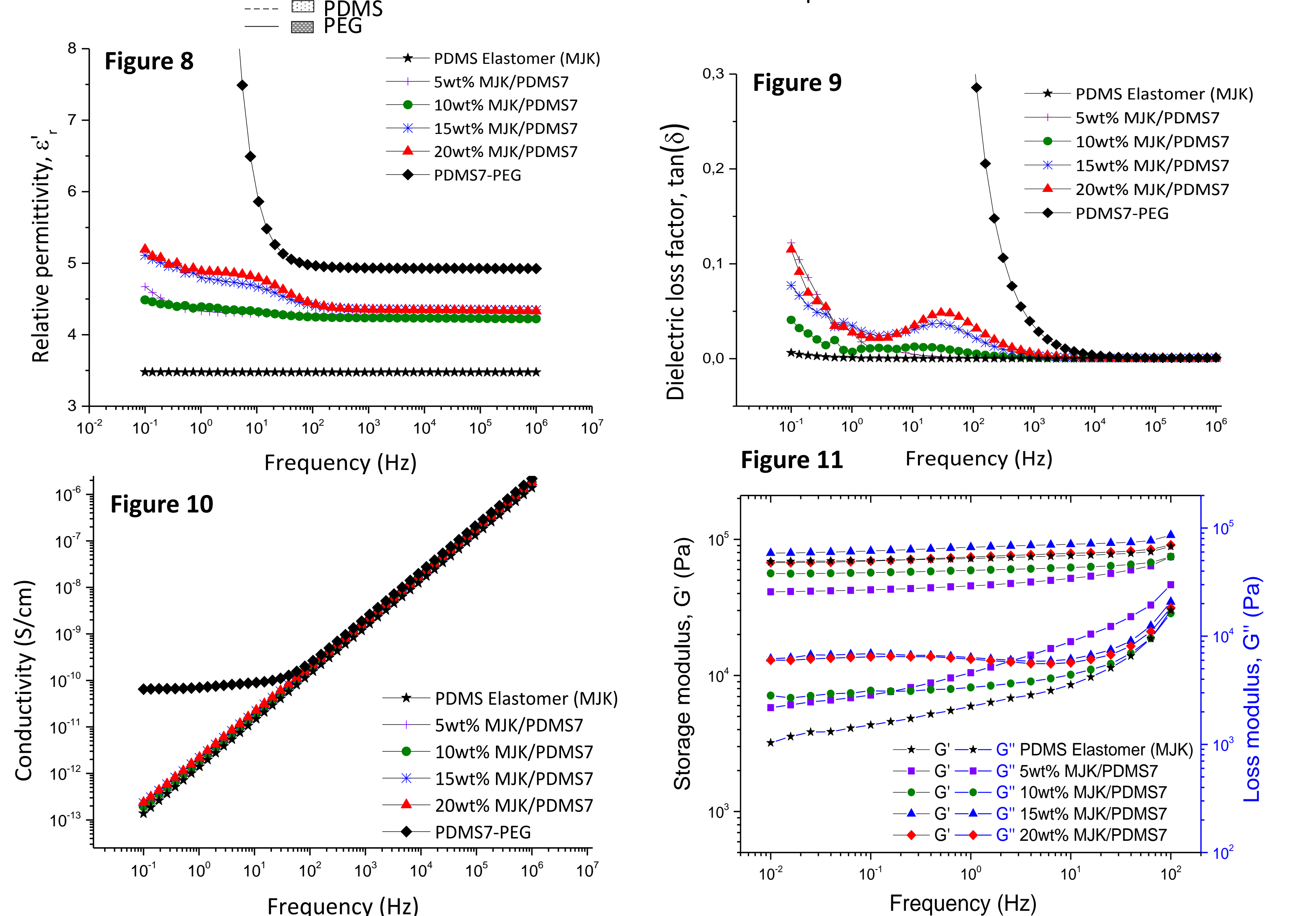


Figure 7: Illustration of morphologies for BPB of PDMS-PEG block copolymer and silicone elastomer: **a.** Continuous phase in PDMS (○: Schematic diagram of chain conformations of PDMS-PEG block copolymer in PDMS matrix) **b.** Co-continuous phase in PDMS **c.** Discontinuous phase in PDMS.



Results for polymer blends MJK/PDMS7 at 23 °C: **Figure 8** Relative permittivity. **Figure 9** Dielectric loss factor. **Figure 10** Conductivity. **Figure 11** Storage and loss modulus.

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Acknowledgements

The Malaysian Ministry of Education (MoE) and Universiti Tun Hussein Onn Malaysia (UTHM) as well as Innovationsfonden Danmark are gratefully acknowledged for their funding.